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Thermodynamic screening of alternative refrigerants for R290 and R600a[☆]

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ABSTRACT

Due to Regulation EU 517/2014 (F-Gas), the use of refrigerants with GWP higher than 150 was limited from 2015 on in small capacity refrigeration systems in Europe. Although refrigerants that dominate are R600a and R290 nowadays, which are classified as A3 by ASHRAE 34, literature points out that some refrigerant mixtures exist that could offer benefits from the point of view of COP and VCC, especially mixtures of hydrocarbons.

This study tries to identify possible mixtures which could be used as alternatives to pure hydrocarbons, with the aim to increase the energetic behaviour of the systems and, if possible, to reduce the flammability characteristics. A thermodynamic screening of refrigerant mixtures (with the base refrigerants R290, R600a, R600, R1270, R152a, R32, R1234yf, R1234ze(E), R1233zd, R744 and R134a) is presented and detailed for two typologies of systems: single-stage systems with adiabatic capillary tube and single-stage systems with non-adiabatic capillary tube. The screening identifies the most promising refrigerant mixtures that will be tested later in experimental systems.

1. Introduction

Energy use of refrigeration and air conditioning systems absorbed about 20% of the total electricity produced in the World and it was responsible of 7.8% of total greenhouse gas emissions in 2014, according to the International Institute of Refrigeration [1]. Inside this sector, the subgroup of domestic fridge/freezers and stand-alone commercial refrigeration appliances, with more than 1.5 billion units, absorbed a 15.4% of the generated electricity, thus they accounted for approximately a 2.6% of electricity produced in the world [2].

To reduce the environmental impact of the refrigeration sector globally, the World and especially the European Union have implemented different agreements and regulations that affect the refrigeration sector, such as the Kigali amendment to the Montreal Protocol [3] or the F-Gas Regulation [4]. However, specifically in relation to the domestic and stand-alone commercial systems and in addition to those mentioned previously, Europe has implemented two important regulations: The Eco-design directive [5] that establishes a minimum level of energy efficiency to be reached by the products that can be placed on the market; and the Energy labelling Regulation ([6] for domestic systems and [7] for commercial systems) which will classify them into efficiency groups.

Obviously, this segment of refrigeration appliances must meet many requirements, such as security, use of low-GWP (low Global Warming Potential) refrigerants and energy efficiency, being sometimes difficult to meet them all simultaneously. Nonetheless, what is clear is that the low-GWP refrigerants (HCs, and HFOs) will be the selected option [8]. Improve security and increase energy efficiency are the pending conditions. In the last two decades, refrigeration manufacturers expanded the use of hydrocarbons as refrigerants for equipment able to operate with maximum refrigerant charge of 150 g, R-600a dominates the domestic sector and R-290 the commercial one [9]. However, the recent modification of the IEC standard [10] will allow to increase the refrigerant charge with A3 refrigerants up to 500 g, thus, its use will be extended to larger systems. In relation to upgrade the performance of these systems, few efforts are being done to improve the performance of the refrigerant. But, literature reflects that there are some options that could enhance energy performance by the use of refrigerant mixtures, such as the mixture R-290/R600a ([11–13]) with energy consumption reduction in refrigerators between 2 and 4%; the use of R-152a [14,15] with energy improvements in a freezer of 7%, or the mixture R-1234yf/R-134a with improvements in relation to R-134a between 14 and 16% [16,17].

Although not considered by the scientific community up to the moment, to the best knowledge of the authors, the search for alternative

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Nomenclature

COP	coefficient of performance
GWP	global warming potential for 100 years (4th AR)
Glide	effective glide in heat exchanger, K
h	specific enthalpy, kJ·kg^{-1}
HC	hydrocarbon
HCFO	hydroclorofluorocarbon
HFC	hydrofluorocarbon
HFO	hydrofluoroolefin
m	mass fraction, %
NBP	normal boiling point, ºC
p	absolute pressure, bar
S	specific entropy, kJ·kg^{-1}
SH	degree of superheating in evaporator, K
SUB	degree of subcooling in condenser, K
t	temperature, ºC
VCC	volumetric cooling capacity, kJ·m^{-3}
x_v	vapour quality

Greek symbols

ε	thermal effectiveness
ν	specific volume, m^3·kg^{-1}

Subscripts

dis	compressor discharge
ihx	internal heat exchanger (non-adiabatic capillary)
in	inlet
K	condensing level
l	saturated liquid
m	average value
O	evaporating level
out	outlet
suc	compressor suction
v	saturated vapour

refrigerants to hydrocarbons has not been considered, but the experimental investigations found in literature indicate that there is room for improvement. Accordingly, the objective of this work is to present the results of a thermodynamic screening of refrigerant mixtures which could be '*better*' refrigerants or at least reduce the energy consumption of stand-alone systems based on R-600a or R-290 and to analyse if thermodynamically the hypothesis is possible. Thus, here, a systematic search based on thermodynamic models has been conducted by considering 120 possible ternary mixtures with 231 combinations in each one for 4 different working conditions, giving a total number of combinations of 110880. This screening has identified a small set of combinations, which, thermodynamically could offer energy benefits. Thus, this is the first stage of a new line of research, which will bring about the scientific community possibilities to test the proposed mixtures in real systems to obtain definite conclusions.

2. Refrigeration typologies and thermodynamic model

Simulations were carried out for the most common refrigeration typologies used in small capacity hermetic refrigeration systems: single-stage systems with adiabatic capillary tube (Fig. 1) and single-stage systems with non-adiabatic capillary tube (Fig. 2). The main difference between them is the heat transfer in the capillary tube. In the first one (Fig. 1), the capillary is alone, whereas in the non-adiabatic layout (Fig. 2) it is wrapped around the suction line, acting as a suction-line to liquid-line heat exchanger [18]. For simulations, both compression and expansion processes were considered as ideal (isentropic and isenthalpic respectively) and pressure drops and kinetic and potential variations were neglected.

The refrigerants were evaluated and compared at fixed evaporating and condensing temperatures for the considered cycles (conditions are presented subsequently in Table 2). Evaporation pressure (p_o), corresponding to evaporating temperature level, was calculated with an iterative method by using the mean enthalpy in the evaporator, as stablished by Eq. (1) and Eq. (2). This criteria is the most recommended for the evaluation of refrigerant mixtures, as suggested by Radermacher and Hwang [19].

$$h_m = \left(\frac{h_{O,in} + h_{O,out}}{2}\right) \tag{1}$$

$$p_O = f(t_O,\ h_m) \tag{2}$$

Condensing pressure (p_k), following the same criteria, was calculated using Eq. (3), that considers that the medium enthalpy matches with 50% vapour quality (x_v) in the condenser.

$$p_k = f(t_k,\ x_v = 0.5) \tag{3}$$

Outlet temperature of evaporator and condenser were evaluated using Eq. (4) and Eq. (5) respectively, considering saturation temperatures at the corresponding pressure and the degree of superheat in the evaporator and degree of subcooling in the condenser.

$$t_{O,out} = f\left(t_{v,p_O} + RU,\ p_O\right) \tag{4}$$

$$t_{K,out} = f(t_{l,Pk} - SUB,\ p_K) \tag{5}$$

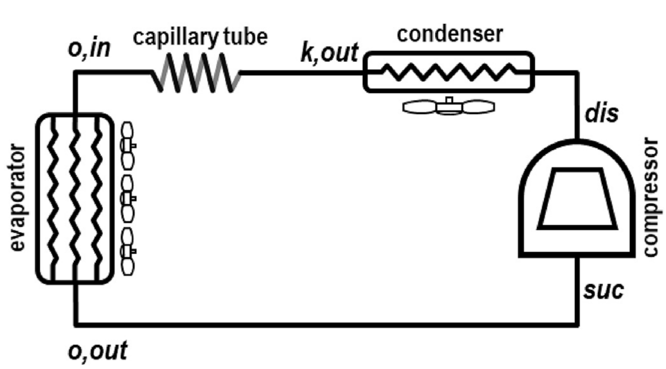

Fig. 1. Single-stage cycle with adiabatic capillary tube.

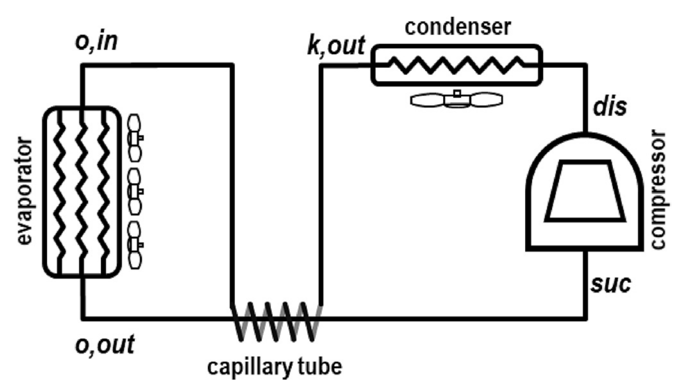

Fig. 2. Single-stage cycle with non-adiabatic capillary tube.

Suction temperature and enthalpy were evaluated considering that the capillary acts as an internal heat exchanger with defined thermal effectiveness, as expressed by Eq. (6) and Eq. (7). Finally, evaporator inlet enthalpy was obtained through the heat balance in the capillary tube.

$$t_{suc} = t_{O,out} + \varepsilon_{ihx} \cdot (t_{K,out} - t_{O,out}) \tag{6}$$

$$h_{suc} = f(t_{suc}, p_O) \tag{7}$$

$$h_{O,in} = h_{K,out} - h_{suc} + h_{O,out} \tag{8}$$

With the thermodynamic states of the refrigerants through the cycles, the following parameters were considered to analyse the performance of the tested refrigerant mixtures:

- **Effective glide**: in evaporator and condenser, the effective glide corresponds to the temperature difference along the phase-change process at constant pressure, as defined by Eq. (9) for the evaporator and by Eq. (10) for the condenser.

$$Glide_O = t(p_O, x_v = 1) - t(p_O, x_{v,O,in}) \tag{9}$$

$$Glide_K = t(p_K, x_v = 1) - t(p_K, x_v = 0) \tag{10}$$

- **Volumetric Cooling Capacity (VCC) and Coefficient of Performance (COP)**, which were evaluated using Eq. (11) and Eq. (12), respectively.

$$VCC = \frac{h_{O,out} - h_{O,in}}{v_{suc}} \tag{11}$$

$$COP = \frac{h_{O,out} - h_{O,in}}{h_{dis,S}(p_K, s_{suc}) - h_{suc}} \tag{12}$$

- **Global warming potential (GWP)** of the mixture, which as evaluated using Eq. (13) as the sum of partial masses of each refrigerant multiplied by their GWP, these values being extracted from the 4th Assessment Report of the IPCC [20].

$$GWP = \sum_i^n (m_i \cdot GWP_i) \tag{13}$$

It is important to mention that the model evaluated the thermodynamic properties of the mixtures using the most recent version of Refprop, version 10 [21], with the recommended mixing rules described in the file HMX.BNC version 4, although described later, in some cases the mixing rules provided incoherent results. Maltab 2016a was the platform used to perform the screening.

3. Boundary conditions and optimization process

Table 1 shows the refrigerants considered in the optimization process and their main properties. All of them have a GWP below 150, thus there is no limitation in the percentage that can be used in the mixture, except for the R-32, that only can be existing in a 22.1% as maximum. According to Ashrae classification [22], all of them (except for the R-744) present flammable characteristics, however R-152a is classified as A2 and R-32, R-1234yf, R-1234ze and R-1233zd as A2L, so depending on the composition, the mixtures can present lower flammability characteristics than isobutane and propane, which are A3.

Two different applications are considered in the optimization process: conservation of fresh product and conservation of frozen product (with selected evaporation temperatures of −10 °C and −30 °C, respectively). For each application, calculations were conducted considering that the capillary was adiabatic or non-adiabatic, as described in Section 2. For the non-adiabatic case (Fig. 2), an 80% thermal efficiency was considered. Condensation temperature, superheating (SH) and subcooling (SUB) were maintained for all cases, taking values of 40 °C, 4 K and 1 K, respectively. The working conditions are summarized in Table 2.

Table 2
Working conditions (M and L refer to “medium” and “low” temperature respectively).

Working conditions	
M1	$t_o = -10$ °C, $t_k = 40$ °C, non-adiabatic (ε = 80%), SH = 4 K, SUB = 1 K
M2	$t_o = -10$ °C, $t_k = 40$ °C, adiabatic, SH = 4 K, SUB = 1 K
L1	$t_o = -30$ °C, $t_k = 40$ °C, Non-adiabatic (ε = 80%), SH = 4 K, SUB = 1 K
L2	$t_o = -30$ °C, $t_k = 40$ °C, Non-adiabatic, SH = 4 K, SUB = 1 K

The purpose of the optimization process is to discover refrigerant mixtures with best energy indicators (COP, VCC) than isobutane (R-600a) and propane (R-290) in the four working conditions considered. For that purpose all possible combinations of three refrigerants of Table 1 were evaluated. Optimization was limited to three components at this moment due to the long-required computation time. For that end, three steps were carried out:

- Finding composition that maximizes COP for each combination of three refrigerants. In this process, the mixtures were formed by varying 5.0% the mass percentage of each fluid.
- Filtering of results and election of mixtures with most suitable properties in terms of COP and VCC.
- Finding composition that maximizes COP for the selected mixtures. The mass variation for each refrigerant involved in the mixture was of 0.5%.

To find the optimal composition, additional restrictions were imposed: To meet the requirements of the EU517/2014 [4], the maximum GWP of the mixture was set at 150. Mixtures exceeding this limit were discarded. The next step was to evaluate the energy parameters of the cycle for each working condition, with the model described in Section 2. As the fluids mixed may have very different Normal Boiling Point (NBP), the glide in the evaporator was a parameter that was controlled and restricted, since a huge glide affects significantly to the

Table 1
Refrigerants considered in the screening and their properties.

	Family	NBP (°C)	Critical temperature (°C)	Critical pressure (bar)	GWP (AR4)	Safety Group
R-290	HC	−42.1	96.7	42.5	3	A3
R-600a	HC	−11.7	134.7	36.3	3	A3
R-600	HC	−0.5	152.0	38.0	3	A3
R-1270	HC	−47.7	92.4	46.6	1.8	A3
R-152a	HFC	−24.0	113.3	45.2	124	A2
R-32	HFC	−51.7	78.1	57.8	675	A2L
R-1234yf	HFO	−29.4	94.7	33.8	4	A2L
R-1234zeE	HFO	−19.0	109.4	36.3	6	A2L
R-1233zd	HCFO	18.3	116.5	36.2	1[a]	A2L
R-744	Natural	−78.4	31.0	73.8	1	A1

[a] Adapted from the 5th AR of the IPCC [23].

thermal performance of the heat exchangers. Therefore, the maximum effective glide allowed in evaporator was of 10 K, although larger values are found in commercial refrigerants [24]. Accordingly, mixtures that surpassed this value were discarded. Another parameter to focus on was the discharge temperature. As it was calculated assuming an isentropic process in the compressor, the restriction value should be moderated. When assuming adiabatic capillary, the limit value was fixed in 70 °C, whereas when non-adiabatic capillary, the value was let free, since it was observed that 70 °C was too restrictive. In that case, the results were analysed afterwards and filtered to discard any nonsense result. This process was carried out for all the possible combinations that each trio of refrigerants can offer. As a result, the combination with better COP was elected. The process is represented in Fig. 3.

4. Screening results

The number of evaluated mixtures were 110880, representing of 120 possible ternary mixtures, 231 combinations for each ternary mixture (5% of mass variation) and 4 different working conditions. Section 4.1 presents the results of the optimization with 5% percentage mass variation, section 4.2 the remaining mixtures when the compatibility restrictions were applied, and section 4.3 the fine optimization of the mixtures that fulfilled the restrictions.

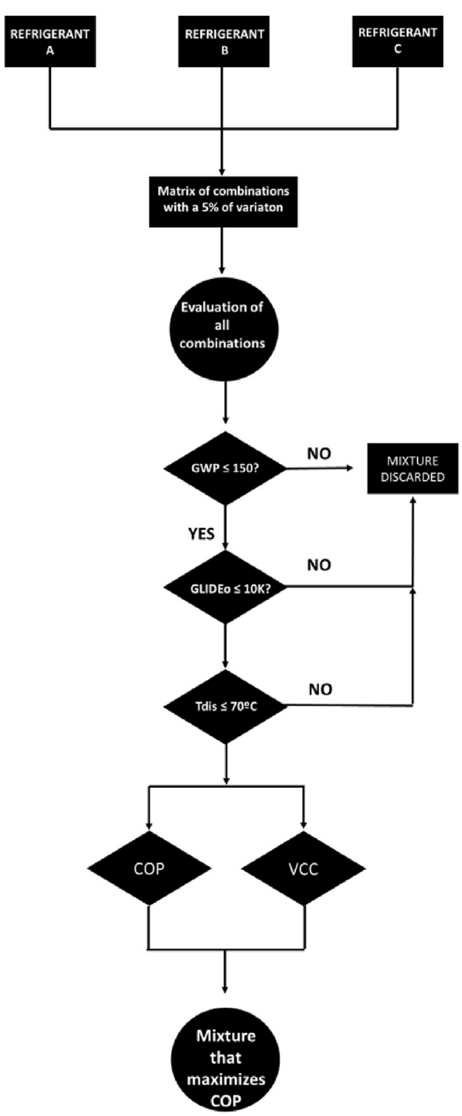

Fig. 3. Scheme of optimization process.

4.1. First optimization process

Fig. 4 and Fig. 5 show, as an example, the result of the first optimization process for M1 and L1 working conditions, which consists in finding the optimal composition for each ternary mixture. As it can be observed, only mixtures with similar or higher COP than the base fluid are considered. Also, it can be observed that with some specific mixtures high COP values were predicted. Obviously, these large values could be associated to a fail of the mixing rules of the components of the database program. Accordingly, the optimal compositions were subjected to an acceptance data range, as explained in Section 4.2.

4.2. Compatibility restrictions

Results obtained for all the possible ternary combinations were compared with reference to the COP and VCC calculated for R-600a and R-290 in each working condition, resulting in four scenarios for each base refrigerant. The considered acceptance range for each scenario was of ±30% of the VCC of the base refrigerant (since for a correct operation of the compressor this value must be similar), and between 0 and +15% of the base refrigerant COP. Higher COP increments were considered as a fail of the mixing rules available in Refprop 10 [21] and therefore the resulting mixtures were not considered as reliable.

Fig. 6 represents all the refrigerant mixtures identified in the first optimization process in relation to R-600a, where it is represented the R-600a operating point in each working condition (Table 2) and the acceptance gap as defined previously. For the operation at medium evaporating conditions (condition M, Table 2), 10 binary and 2 ternary mixtures fulfilled the restrictions, with maximum COP increments in relation to R-600a up to 6.0% and variations in the VCC between −24.5

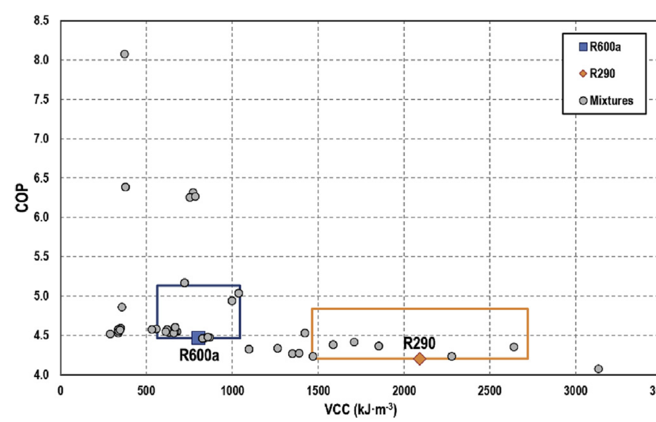

Fig. 4. Resulting mixtures with t_O = -10 °C with non-adiabatic capillary.

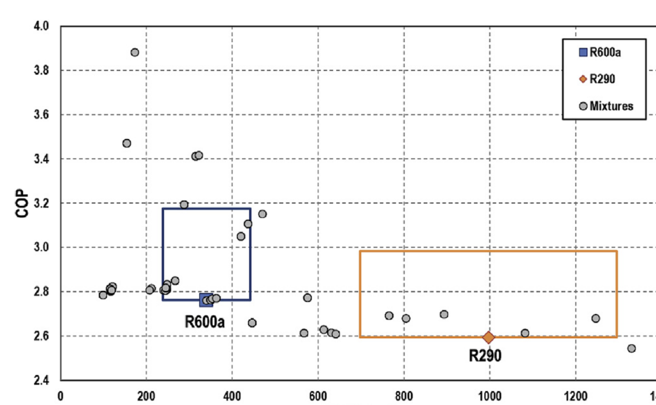

Fig. 5. Resulting mixtures with t_O = -30 °C with non-adiabatic capillary.

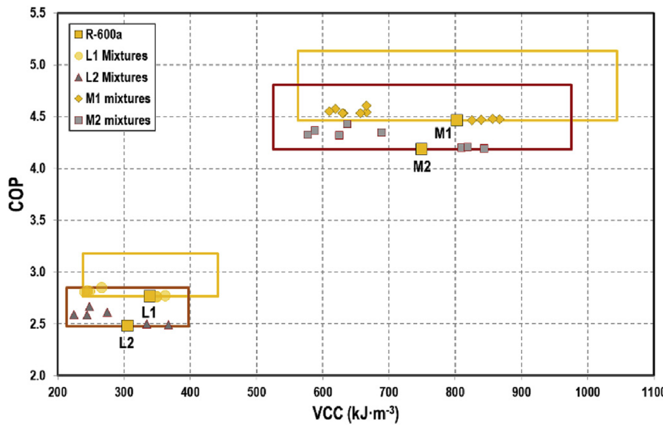

Fig. 6. Alternative mixtures in the acceptance gap for R-600a.

and 11.5%. At low evaporating levels (condition L, `), only 8 binary blends were identified, with maximum COP increment up to 8.6% and variations in the VCC between −28.0% and 14.2%.

Fig. 7 depicts the refrigerant mixtures identified in the first process with R-290 as base refrigerant, as well as, its operating point and acceptance gap at each working condition. At medium temperature (condition M, Table 2), 5 binary and 2 ternary mixtures and 1 pure fluid were identified. R-152a, with reduction in VCC below −30% was also considered because it provided an interesting COP increment. Maximum COP increments reached 11.3% with variations in VCC between −24.5 and 11.5%. At low evaporating level (condition L, Table 2). 6 binary and 1 ternary mixture were in agreement with the restrictions, as well as, one pure fluid. Additionally, R-744/R-290 blend and R-1234yf were also included because they were at the limit of the acceptance gap. For this condition, maximum COP increment was of 11.6% and the variation of the VCC was between −27.5 and 25.8%.

4.3. Fine optimization process

Finally, mixtures identified in the first optimization process that fulfilled the compatibility restrictions were subjected to a fine optimization process, with the same calculation procedure described in Section 2, but considering 0.5% of mass fraction variation of each component.

4.3.1. Alternative mixtures to R-600a

Figs. 8-11 represent the thermodynamic properties of alternative mixtures to R-600a in the four operating conditions considered (Table 2). It can be seen that the identified mixtures are mostly binary mixtures, mainly composed by R-600a (its composition varies between 96.5% and

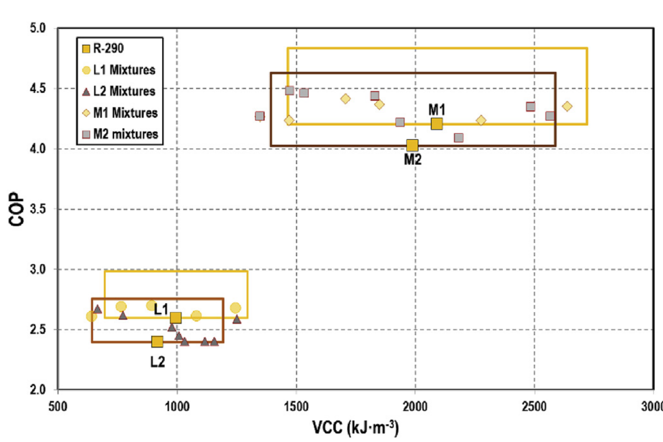

Fig. 7. Alternative mixtures in the acceptance gap for R-290.

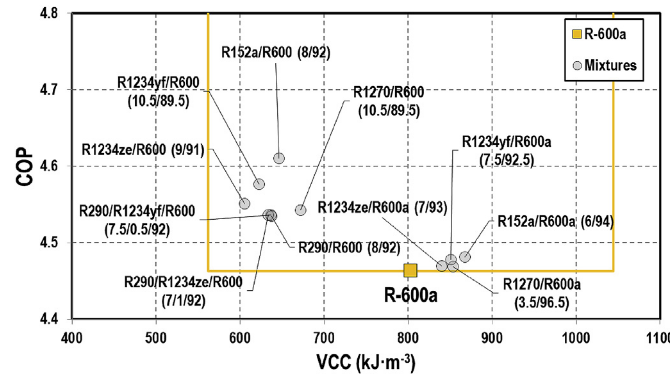

Fig. 8. Mixtures in the acceptance gap for R-600a in M1 conditions.

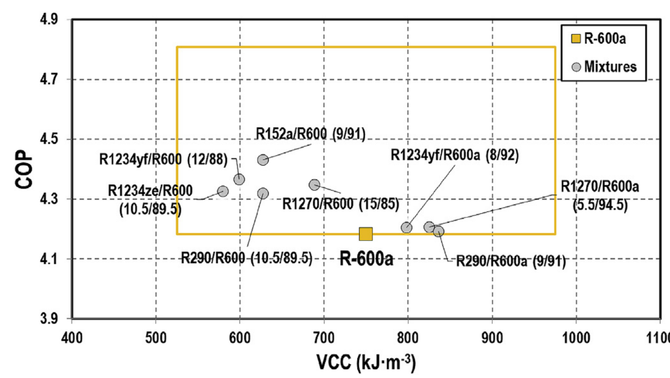

Fig. 9. Mixtures in the acceptance gap for R-600a in M2 conditions.

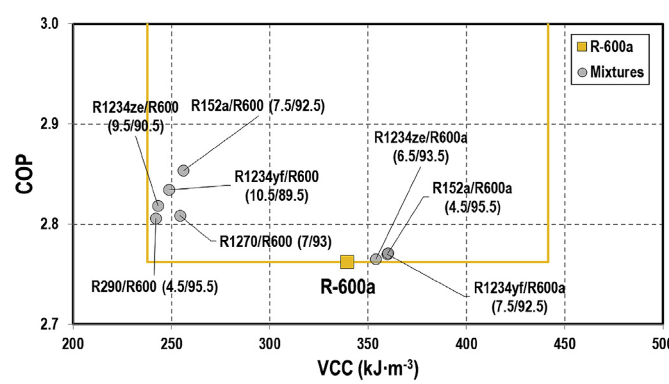

Fig. 10. Mixtures in the acceptance gap for R-600a in L1 conditions.

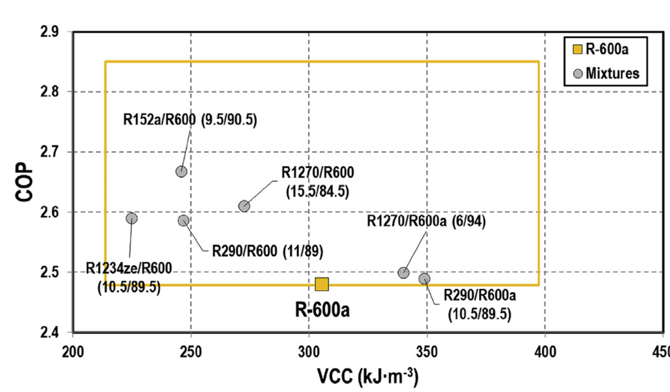

Fig. 11. Mixtures in the acceptance gap for R-600a in L2 conditions.

89.5%) or R-600 (between 95.5% and 84.5%) with a small fraction of another fluid, although there are two ternary blends with a small fraction of a third component (below 1%). All mixtures with R-600a achieve a slight increment in COP (between 0.1% and 0.4% for M1, 0.6% and 0.3% for M2, 0.3% and 0.1% for L1 and 0.8% and 0.4% for L2) and higher VCC (between 4.6% and 8% for M1, 6.4% and 11.5% for M2, 4.2% and 6% for L1 and 11.2% and 14.2% for L2). On the contrary, the mixtures composed by R600 achieve higher increments in COP (between 3.3% and 1.6% for M1, 6% and 3.3% for M2, 8.6% and 1.7% for L1 and 7.6% and 4.3% for L2), but the VCC decreases significantly (between −24.7% and −16.4% for M1, -22.7% and −16.4% for M2, -28.7% and −24.6% for L1 and -26.4% and −10.8% for L2). Another aspect to consider is the glide in the phase change processes, which is accentuated when the difference between the NBP of the refrigerants is greater, being higher with R-600.

4.3.2. Alternative mixtures to R-290

Figs. 12-15 highlight the mixtures alternative to R-290 identified for all the operating conditions (Table 2). In this case the number of mixtures is shorter than for R-600a. For propane alternatives, two types of mixtures can be observed: mixtures composed by a low percentage of R-744 and another fluid (the R-744 increases the VCC of the mixture), and those constituted by R-290 as the main component with another refrigerant with low percentage. COP increases up to 5.1% in M1 conditions, 11.7% in M2, 4.1% in L1 and 11.6% in L2. It must be considered that the mixtures with R-744 have a significant glide, being around 10 K in the evaporator, which could lead into operational problems. It is important to mention that the pure fluids R-152 and R-1234yf are in the limit of the acceptance range. These fluids will provide small increments in COP in most of the cases but with a strong reduction in capacity.

5. Conclusions

A theoretical search for refrigerant mixtures that could provide energy improvements in relation to R-600a and R-290 for refrigeration systems was performed. Blends with a maximum of three components were subjected to a thermodynamic optimization process consisting on finding the optimal composition that maximizes COP subjected to different compatibility restrictions. The screening was focused on the application to two evaporating levels (−10 °C and −30 °C) for constant condensing temperature (40 °C) for two types of typologies: single-stage systems with adiabatic capillary tube and single stage systems with non-adiabatic capillary tube.

In total, 110880 different refrigerant mixtures were evaluated to select the best candidates. Selection chose COP as optimizing parameter, but the blends were subjected to fulfil some compatibility restrictions that guarantee the experimental verification in real systems. Concretely, all the selected mixtures presented a GWP value below 150, operate with an effective glide in the evaporator of 10 K and present an isentropic

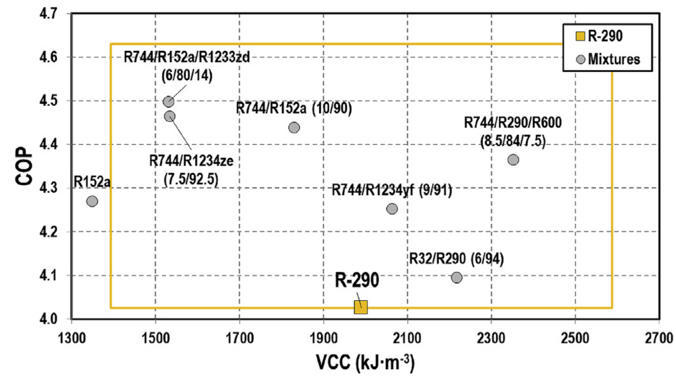

Fig. 13. Mixtures in the acceptance gap for R-290 in M2 conditions.

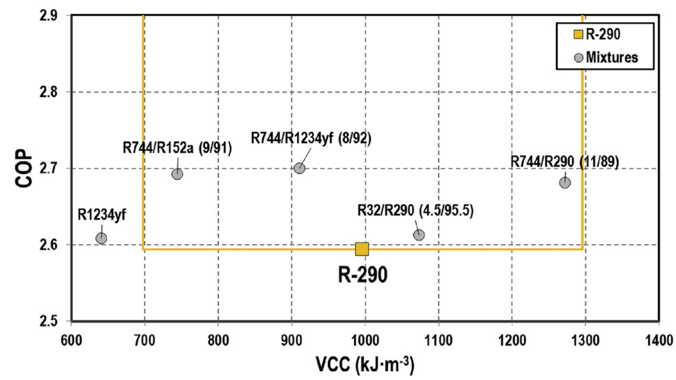

Fig. 14. Mixtures in the acceptance gap for R-290 in L1 conditions.

discharge temperature below 70 °C. From all mixtures fulfilling restrictions, the ones presenting theoretical COP increments from 0 to 15% and variations in the volumetric cooling capacity (VCC) from −30 to 30% in relation to R-600a and R-290 were selected. Finally, remaining mixtures were optimized again with a mass fraction variation of each component of 0.5%.

Respect to possible alternatives to isobutane (R-600a), mixtures composed by R-1234yf/R-600a and R-1270/R-600a offer a slight increase in COP (between 0.3% and 0.6% and between 0.1% and 0.8% respectively) and a small increment in VCC (between 5.9% and 6.4% and between 6.3% and 11.2% respectively) respect R-600a, whereas mixtures form by R-1270/R-600, R-152a/R-600, R-1234zeE/R-600 and R-290/R-600 achieve high increments of COP (between 1.7% and 5.3%, 3.3% and 7.6%, 2.5% and 4.4%, 2% and 4.5% and 1.6% and 8.6%) but the VCC is decreased significantly (between −25.1% and −8.2%, −24.6% and

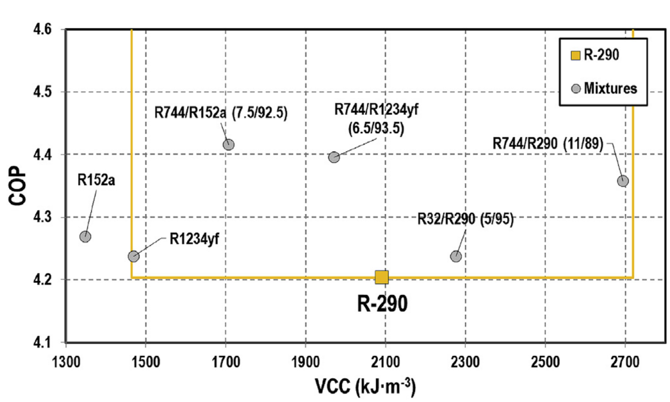

Fig. 12. Mixtures in the acceptance gap for R-290 in M1 conditions.

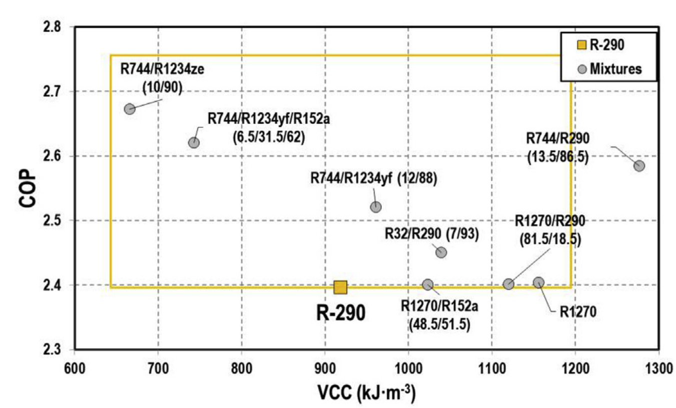

Fig. 15. Mixtures in the acceptance gap for R-290 in L2 conditions.

−16.4%, −24.6% and −16.4%, −28.4% and −22.7% and −28.4% and −16.4%, respectively) respect R-600a.

Respect to possible alternatives to propane (R-290), mixtures form by a small proportion of R-744 with R-290, R-1234yf, R-152a or R-1234ze(E) achieve high increments of COP (between 3.4% and 7.9%, 4.1% and 5.6%, 3.8% and 10.3% and 10.9% and 11.6%, respectively) but the VCC differs a lot between one mixture with each other (from 28.8% to 38.9%, −8.6% and 3.7%, −25.4% and −8%, −27.6% and −23% and 7.7% and 13%, respectively). It is also identified the mixture R-32/R-290, which accomplishes a raise of COP and VCC between 0.8% and 2.3% and 8.8% and 13%, respectively.

Accordingly, from the theoretical screening performed in this work, it is clear that there are some refrigerant mixtures which could be candidates to replace pure hydrocarbons in small systems, providing a slight increment in COP. This work constitutes an initial hypothesis, since experimental validation is needed to confirm the real possibilities of the identified mixtures.

Author contribution

DC and RL conceived the idea, performed the theoretical simulations. LN, JC, DS and RC checked the validity of the results and contributed to the selection of refrigerant mixtures. The manuscript was written by DC and RL and revised by LN.

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